

# Tunneling Recombination in Optically Pumped Graphene with Electron-Hole Puddles

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We evaluate recombination of electrons and holes in optically pumped graphene associated with the interband tunneling between electron-hole puddles and calculate the recombination rate and time. It is demonstrated that this mechanism can be dominant in a wide range of pumping intensities. We show that the tunneling recombination rate and time are nonmonotonic functions of the quasi-Fermi energies of electrons and holes and optical pumping intensity. This can result in hysteresis phenomena.

The gapless energy spectrum of electrons and holes in graphene layers (GLs) and non-Bernal stacked multiple graphene layers (MGLs) [1–3], which leads to specific features of its electrical and optical processes, opens up prospects of building of novel devices exploiting the interband absorption and emission of terahertz (THz) and infrared (IR) photons. Apart from an obvious possibility to use of graphene in THz and IR photodetectors [4–6], graphene layers (GLs) and multiple graphene layers (MGLs) can serve as active media in THz or IR lasers with optical [7–12] and injection [13] pumping. The achievement of sufficiently strong population inversion, which is necessary for negative dynamic conductivity in the THz or IR ranges of frequencies, can be complicated by the recombination processes. The recombination of electrons and holes in GLs at not too low temperatures is mainly determined by the emission of optical photons [14]. Due to a relatively high energy of optical phonons ( $\hbar\omega_0 \simeq 0.2$  eV), the recombination rate in GLs associated with this mechanism can be acceptable even at the room temperatures [10–12]. The radiative recombination in the practically interesting temperatures is weaker than that due to the optical phonon emission [15]. The acoustic phonon recombination mechanism as well as the Auger mechanism are forbidden due to the linearity of the electron and hole spectra even in the case their modifications associated with the inter-carrier interaction [16, 17]. More complex processes involving the electron and hole scattering on impurities assisted by acoustic phonons also provide rather long recombination times which are longer than the radiative recombination time [18]. However in GLs with a disorder caused, for instance, by fluctuation of the surface charges resulting in the formation of the electron and hole puddles [19] (see also, for instance, Refs. [20–22]), the recombination can also be associated with the interband tunneling in the spots where the build-in fluctuation field is sufficiently strong.

In this paper, we find the dependences of the recombination rate and time in graphene with electron-hole puddles on the quasi-Fermi energy of electrons and holes (pumping intensity). Our results can be used for the interpretation of experimental observations and to promote the realization of graphene-based THz and IR lasers.

The electric potential  $\varphi = \varphi(x, y, z)$ , where  $x$  and  $y$  are the coordinates in the GL plane and the coordinate  $z$  is directed perpendicular to this plane, is governed by the Poisson equation presented in the following form:

$$\Delta\varphi = \frac{4\pi e}{\epsilon}(\Sigma_e - \Sigma_h - \Sigma_i) \cdot \delta(z). \quad (1)$$

Here,

$$\Sigma_e = \frac{2}{\pi\hbar^2} \int_0^\infty \frac{dp p}{1 + \exp\left(\frac{v_W p - \mu_e + e\varphi}{k_B T}\right)},$$

$$\Sigma_h = \frac{2}{\pi\hbar^2} \int_0^\infty \frac{dp p}{1 + \exp\left(\frac{v_W p + \mu_h - e\varphi}{k_B T}\right)},$$

and  $\Sigma_i$  are the electron, hole, and charged impurity sheet densities, respectively,  $e = |e|$ ,  $\epsilon$ ,  $\hbar$ , and  $k_B$  are the electron charge, dielectric constant, reduced Planck's constant, and Boltzmann's constant,  $v_W \simeq 10^8$  cm/s is the characteristic velocity,  $p$  is the electron and hole momentum,  $\mu_e$  and  $\mu_h$  are the electron and hole quasi-Fermi energies counted from the Dirac point,  $T$  is the temperature, and  $\Delta$  is the three dimensional Laplace operator. The delta function  $\delta(z)$  reflexes the localization of electrons, holes, and impurities in the GL plane  $z = 0$ . In Eq. (1), we assumed that the electron spectra or electrons and holes are linear:  $\epsilon_e = v_W p$  and  $\epsilon_h = -Wp$ . In the equilibrium, i.e., in the absence of pumping,  $\mu_e = \mu_h = \mu_i$ , where  $\mu_i$  is determined by the spatially averaged impurity density  $\langle \Sigma_i(x, y) \rangle$  with  $\mu_i = 0$  if the latter averaged value is equal to zero (the Fermi levels correspond to the Dirac point) or by the gate voltage (in gated GL structures). In the case of pumping of intrinsic GLs,  $\mu_e = -\mu_h = \mu \neq 0$ . In the latter case,  $T$  can differ from the lattice temperature  $T_l$  being both higher than  $T_l$  or lower [23].

Considering in the following the case  $\langle \Sigma_d(x, y) \rangle = 0$  and assuming that the fluctuations are not too strong ( $e|\varphi| < k_B T$ ), Eq. (1) can be linearized:

$$\Delta\varphi = \left[ \frac{\varphi}{r_s} \ln\left(1 + e^{\mu/k_B T}\right) - \frac{4\pi e}{\epsilon} \Sigma_i \right] \cdot \delta(z), \quad (2)$$

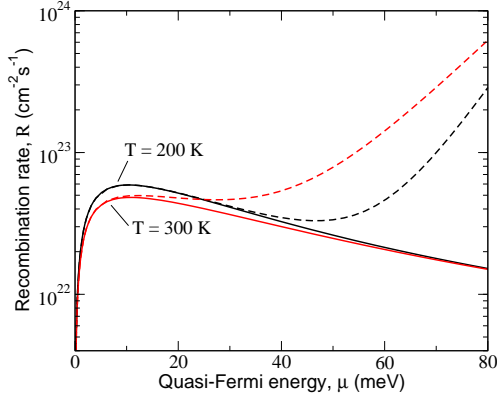


FIG. 1: (Color online) Recombination rate  $R$  vs quasi-Fermi energy  $\mu$  for tunneling mechanism (solid lines) and for combination of tunneling and optical phonon mechanism (dashed lines) in a GL at different temperatures  $T$ .

where  $r_s = (\hbar^2 v_W^2 / 16 e^2 k_B T)$  is the screening length.

Solving Eq. (2) with the boundary conditions  $\varphi|_{z=\pm\infty} = 0$  assuming that

$$\Sigma_i = \sum_{q_x, q_y} \Sigma_{q_x, q_y}^{(i)} \exp[i(q_x x + q_y y)],$$

$$\varphi(x, y, z) = \sum_{q_x, q_y} \Phi_{q_x, q_y} \exp(-q|z|) \exp[i(q_x x + q_y y)],$$

where  $\Sigma_{q_x, q_y}^{(i)}$  and  $\Phi_{q_x, q_y}$  are the pertinent amplitudes and  $q = \sqrt{q_x^2 + q_y^2}$ , for the amplitude of the potential at the GL plane we obtain

$$\Psi_{q_x, q_y} = \frac{2\pi e}{\hbar \left[ q + \frac{1}{2r_s} \ln(1 + e^{\mu/k_B T}) \right]} \Sigma_{q_x, q_y}^{(i)}. \quad (3)$$

Setting  $q \sim \pi/\bar{a}$ , where  $\bar{a}$  is the characteristic size of the puddles, and using Eq. (3), one can express  $\Psi = \max|\Psi_{q_x, q_y}|$  via its value in the absence of screening  $\bar{\Psi} = 2e\bar{a}\bar{\Sigma}/\hbar$ :

$$\Psi = \frac{\bar{\Psi}}{\left[ 1 + \frac{\bar{a}}{2\pi r_s} \ln(1 + e^{\mu/k_B T}) \right]}. \quad (4)$$

According to Eq. (4), the fluctuation electric field between positively and negatively charged puddles can be estimated as  $\mathcal{E} \sim \Psi/\bar{a}$ .

Using the general formulas for the probability of the interband tunneling in GLs [24–26] and considering Eq. (4), the rate of the tunneling recombination can be presented as

$$R \sim \frac{2\sqrt{2}e^{3/2}}{\pi^2 \hbar^{3/2} v_W^{1/2}} \sqrt{\frac{\bar{\Psi}}{\bar{a}}} \left( \frac{\mu}{e\bar{a}} \right) = \frac{\bar{R}(\mu/e\bar{\Psi})}{\sqrt{1 + \eta_s \ln(1 + e^{\mu/k_B T})}}, \quad (5)$$

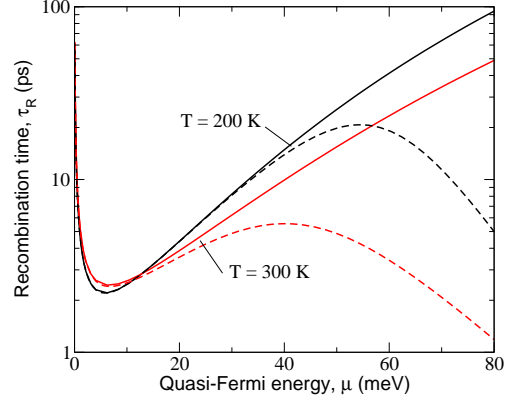


FIG. 2: (Color online) Recombination time  $\tau_R$  vs quasi-Fermi energy  $\mu$  at different temperatures  $T$ . Solid lines correspond to tunneling recombination, while dashed lines correspond to recombination associated with both tunneling and phonon mechanisms.

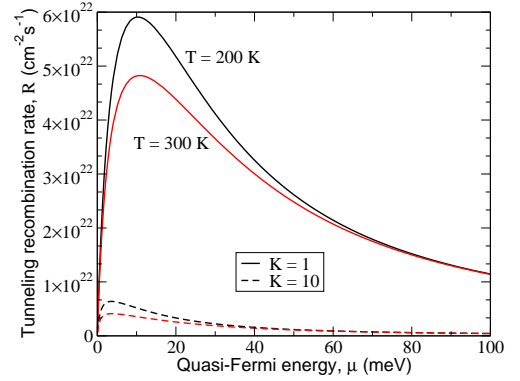


FIG. 3: (Color online) Tunneling recombination time  $\tau_R$  vs quasi-Fermi energy  $\mu$  at different temperatures  $T$  in a GL (solid lines) and in MGL structure with ten GLs, i.e.,  $K = 10$  (dashed lines).

$$R \sim \frac{2\sqrt{2}e^{3/2}}{\pi^2 \hbar^{3/2} v_W^{1/2}} \left( \frac{\Psi}{\bar{a}} \right)^{3/2} = \frac{\bar{R}}{[1 + \eta_s \ln(1 + e^{\mu/k_B T})]^{3/2}} \quad (6)$$

at relatively weak ( $\mu < e\Psi$ ) and strong ( $\mu > e\Psi$ ) optical pumping, respectively. Here,

$$\bar{R} \sim \frac{2\sqrt{2}e^{3/2}}{\pi^2 \hbar^{3/2} v_W^{1/2}} \left( \frac{\bar{\Psi}}{\bar{a}} \right)^{3/2}, \quad \eta_s = \frac{\bar{a}}{2\pi r_s} = \frac{16}{\pi \hbar^2} \frac{e^2 \bar{a} k_B T}{v_W^2}.$$

Assuming that  $\hbar = 4$ ,  $\bar{a} = 30$  nm, and  $\bar{\Sigma} = 4 \times 10^{10} \text{ cm}^{-2}$  [19], for the amplitude of the potential profile fluctuations  $e\bar{\Psi}$  and for the characteristic rate of the tunneling recombination  $\bar{R}$ , we obtain the following estimates:  $e\bar{\Psi} = 8.5$  meV and  $\bar{R} \simeq 2.7 \times 10^{23} \text{ cm}^{-2} \text{ s}^{-1}$ , respectively. At  $T = 77 - 300$  K, one obtains  $r_s \simeq 0.67 - 2.62$  nm. These values are used in the following.

In the case of strong pumping, the effective temperature of the electron-hole plasma can be much higher than the lattice temperature [23]. As a result, the

electron-hole plasma can become nondegenerate, so that  $\mu_e = \mu < 0$  while  $\mu_h = -\mu > 0$ . This is possible when the plasma density increases with increasing pumping intensity slower than the effective temperature rises. In such a case, the screening vanishes ( $\Psi \simeq \bar{\Psi}$ ), and  $R < 0$ . This implies that when  $\mu$  changes its sign, the tunneling recombination of electron-hole pairs turns to their tunneling generation. This is because when the plasma density is lower than it would be in equilibrium but at the effective temperature  $T$ , the tunneling generation tends to establish an equilibrium density corresponding to  $T$ .

The dependences of the tunneling recombination rate  $R$  versus the quasi-Fermi energy  $\mu$  for different temperatures  $T$  are shown in Fig. 1. For the calculations we used a formula for  $R$  which interpolates the dependences given by Eqs. (5) and (6). Figure 1 demonstrates also the dependences of the recombination rate  $R + R_0$  associated with both tunneling and optical phonon mechanisms (dashed lines). The contribution of the optical phonon mechanism  $R_0$  is taken into account using a simplified analytical formula [10, 23] derived from more rigorous one [14]:

$$R_0 = \bar{R}_0 \left[ (\mathcal{N}_0 + 1) \exp\left(\frac{2\mu - \hbar\omega_0}{T}\right) - \mathcal{N}_0 \right] \\ \simeq \bar{R}_0 \exp\left(-\frac{\hbar\omega_0}{k_B T}\right) \left[ \exp\left(-\frac{2\mu}{k_B T}\right) - 1 \right]. \quad (7)$$

Here  $\bar{R}_0 \simeq 10^{23} \text{ cm}^{-2} \text{ s}^{-1}$  [14] is the pertinent characteristic recombination rate and  $\mathcal{N}_0 = [1 + \exp(\hbar\omega_0/k_B T)]$  is the number of optical phonons. Disregarding the effects of electron-hole heating and cooling and the effect of optical phonon heating [23], we have neglected the difference in the effective temperature  $T$  and the lattice temperature  $T_l$ . One can see that  $R$  is a nonmonotonic function of  $\mu$  with a maximum at a certain value of  $\mu$ . This is attributed to an increase in  $R$  at small  $\mu$  due to an increase in the electro and hole densities, i.e. in the number of carriers participating in the tunneling processes. However, at higher values of  $\mu$ , the screening of the potential fluctuations leads to a decrease in the electric field between the puddles and, consequently, in a decrease in the tunneling probability. When  $\mu$  increases further, the tunneling mechanism gives way to the optical phonon mechanism which becomes dominating. This results in a dramatic rise of  $R + R_0$  in the range of large  $\mu$ . The quantity  $\mu$  is governed by an equation equalizing the rate of recombination  $R + R_0$  and the rate of carrier photogeneration  $G = \beta I$ , where  $\beta = 0.023$  is the GL absorption coefficient and  $I$  is the photon flux. Hence, the  $\mu - I$  dependence

can exhibit the S-shape (see the dashed line in Fig. 1 corresponding to  $T = 200 \text{ K}$ ) leading to a hysteresis which might be pronounced at lowered temperatures.

The recombination time is given by

$$\tau_R = \frac{\langle (\Sigma_e + \Sigma_h) \rangle}{2(R + R_0)} \\ = \frac{2}{\pi(R + R_0)} \left( \frac{k_B T}{\hbar v_W} \right)^2 \int_0^\infty \frac{d\varepsilon \varepsilon}{1 + \exp(\varepsilon - \mu/k_B T)}. \quad (8)$$

The recombination time calculated without considering optical phonon mechanisms (solid lines) and with the latter (dashed lines) is shown in Fig. 2. As follows from Fig. 2, the  $\tau_R - \mu$  dependence is also nonmonotonic with a minimum at moderate  $\mu$  (where  $\tau_R \sim 2 \text{ ps}$ ) and a maximum at rather high  $\mu$  (where  $\tau_R \sim 5 - 20 \text{ ps}$ ).

In optically pumped MGL structures, the electron and hole densities in each GL are close to each other provided the number of GLs  $K$  is not too large [27]. If the fluctuations are associated with the charges located near the lowermost GL, say, near the interface between this GL and the substrate, these fluctuations are screened by all GLs. In cases of such MGL structures, the first two terms in the right-hand side of Eq. (1) can be multiplied by a factor  $K$ , so that the screening length becomes much shorter:  $r_s^{(K)} = (\epsilon \hbar^2 v_W^2 / 16 K e^2 k_B T)$ . Consequently, parameter  $\eta_s$  should be substituted by  $K\eta_s$ . Naturally, this leads to a stronger suppression of the fluctuations (by a stronger screening) and, hence to weakening of the tunneling recombination under consideration as shown in Fig. 3. If each GLs contains its own fluctuation charges, the averaging over a number of GL leads to a significant smoothening of the potential relief in all GL and, hence, to the suppression of the tunneling recombination in such MGL structures.

In conclusion, we calculated the recombination and time as functions of the quasi-Fermi energy in optically pumped graphene with electron-hole puddles. It was shown that the tunneling recombination can be dominant recombination mechanism at low and moderate values of quasi-Fermi energy and, hence, at low and moderate pumping intensities. The dependences of the recombination and time on the quasi-Fermi energy and pumping intensity can be nonmonotonic resulting in hysteresis phenomena. The tunneling recombination can be diminished in MGL structures.

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